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Since this AFOSR grant started on January 1, 1999, a number of new results were obtained by this P1 and his group in the field of nonlinear optics and quantum electronics, in the following directions:

- 2.i. Continuing pioneering research on sub-femtosecond pulses and physics with them, in particular 6-ionization by super-short pulses and discovery of so called area resonances in ionization of quantum wells by sub-cycle field pulses
- 2.ii. Pilot pioneering research on nuclear gamma optics, in particular three-photon photon-nuclear fission
- 2.iii. Continuing pioneering collaborative research on fundamental new phenomenon of multi-mode interference in quantum mechanics, in particular highly regular pattern formation by 8-excited wave functions.
- 2.iv. Discovery and study of novel effect of field-gradient-induced second-harmonic generation in nonlinear optics of vacuum.
- 2.v. Preliminary research on new perspective directions

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**New Principles of Laser-Based Radiation and Particle Sources**

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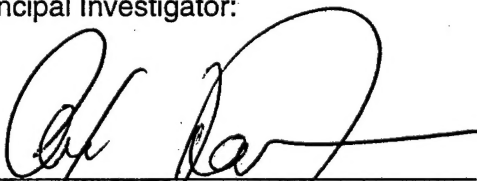
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### *1. Brief review of the PI's work under the AFOSR support*

The research by this principal investigator and his group is done under the AFOSR Grant #F49620-99-1-0096 (E51-2048). This grant was activated on January 1, 1999, with the project period of 33 months ending on September 30, 2001, and the no-cost extension for one more year. By the end of 2001, the research of this PI and his group has been supported by AFOSR continuously for about 22 years. During that period, under AFOSR support, this PI and his group authored or co-authored about 300 publications, among them 10 books and book contributions, 88 regular journal papers, one patent, and 29 conference proceedings; the rest are conference papers.

In particular, under this AFOSR grant (33 months by the end of the grant), 21 new papers have been published [1-21], and two more papers [22-23] have been submitted (and a few more were submitted before the end of the no-cost-extension); this PI is also in the process of a book preparation [24] (with about 2/3 of the project being ready).

All of the effects proposed under the AFOSR support are novel and have initiated new opportunities in the field. The work by this group is highly credited by the research community. Within the last ten years, for example, the work by this PI and his group was cited for about 250 times/year (according to "Science Citation Index ") by other researchers.

One of the most recent research breakthroughs in "extreme" nonlinear optics, was the first experimental observation of atto-second, or more precisely sub-femtosecond ( $\sim 0.25\text{fs}$ ) pulses [25] in June'01. This phenomenon was envisioned in our research under AFOSR support beginning from the pioneering prediction of high-intensity sub-femtosecond pulses in multi-cascade stimulated Raman scattering in 1994, and later on [26]. as well in our prediction of the so called EM-"bubble" pulses [27], that was followed by our theory of 3D-spatial propagation of these super-short pulses [28]. All of these contributions have been highly cited by the researchers in the field. Together with the research of other groups, in particular P. B. Corkum, S. E. Harris, H. C. Kapteyn and M. M. Murnane, and others [29], this work prompted the race to obtain and observe sub-femtosecond pulses, resulting finally in the experiment [25]. Our pioneering work [26] was prominently cited in [25].

Many other, most of them pioneering, contributions by this PI and his group has been at the root of many new research direction in nonlinear optics, EM-physics and laser physics that are current or just getting into experimental or even development phase. The very first, pioneering prediction and experimental observation of the very-high-order *sub-harmonic* oscillations in nonlinear parametric oscillators and based on them frequency dividers by this PI in the beginning of 1960-th [30], are still cited by many researchers. Even leaving aside that work, another earlier contribution, specifically the first prediction of anomalously huge absorption (up to 50%) of micro-wave radiation by very thin ( $\sim 20 - 30\text{\AA}$ ) metallic films made also by him in the beginning of 1960-th [31], has been very recently revived and cited by Dr. E. Yablonovitch and his group [32] in their research on the EM-properties of multi-layered metallic structures.

The prediction by this PI of the so called "*self-bending*" effect by the laser beam with strongly asymmetrical profile in 1968 and later work [33] in the materials with nonlinear refractive index, has been almost immediately verified experimentally, and since then has a long record (including some recent work) of follow-up experimental and theoretical research by other workers. In fact, This PI and his then student, G. Swartzlander, were first in 1989 [33] to



observe for the first time the continuous-wave self-bending effect using sodium vapor as a nonlinear medium.

This PI's earlier work [34], 1969, on the so called *external self-focusing* by a thin nonlinear layer became a precursor of the well developed by now by Dr. Eric Van-Stryland's group the so called *z-scan* method of measuring the nonlinear index of refraction of various materials by measuring the focal distance of external self-focusing; this work of 1969 is still cited and used by the workers in the field. Furthermore, it became a basis for the new effect of optical bistability based on self-focusing [35], predicted and experimentally observed in 1981 in collaborative effort by Bell Lab group (J. E. Bjorkholm, P. W. Smith, and W. J. Tomlinson) and this PI; this effect was the very first observed *cw* optical bistability; besides it was the first one without resonators or nonlinear interfaces, and these results were used later on by many other researchers.

Back in 1969 and the beginning of 1970-ties, this PI and his group [36] developed a broad and detailed theory of nonlinear resonant interaction (including multi-photon interactions such as stimulated Raman) of light and matter, first of all the nonlinear polarizability and nonlinear refractive index, and self-action (self-focusing and self-bending) effects resulting from these interactions. These results became a long-lasting contributions, and together with the book [37], they were used and cited by great many researchers including very recent work.

In 1976 and in his later work [38], this PI made a prediction of hysteretic, bistable, and switching effects in the reflection and refraction of intense light at the interface between linear and nonlinear material, or the so called *nonlinear interfaces*. Very soon, in 1979, based on that prediction, most of these effects have been first time experimentally observed [39] by P. W. Smith, W. J. Tomlinson and their group. There were many novel aspects of the problem, from the very unusual nonlinear Goose-Haenken effect [40] to novel hybrid switching devices [41]; most of them have been reviewed in [42]. The phenomenon of reflection and refraction at nonlinear interfaces became rich and exciting field, and there were many hundreds citations of these pioneering works; the field is still current.

In 1981 and in their later work [43], this PI with P. Meystre were first to predict *self-induced non-reciprocity* of counter-propagating waves in an initially "normal" yet nonlinear medium, and point out a few related device applications, such as huge enhancement of the Sagnac effect in optical gyroscopes due to nonlinearly induced non-reciprocity, and directionally asymmetrical bistability in a symmetrically pumped nonlinear ring interferometer. These results have been used and cited by many other researchers.

The further exploration of the theory of nonlinear counter-propagating waves by this PI and his then student C. T. Law revealed many novel feature of the physics of the phenomenon, in particular interplay of different polarizations of these waves that resulted in *eigen-polarizations*, multi-stability of steady-state modes, etc [45]. A new phenomenon predicted by their theoretical work [46] (and verified later on in experiments by Boyd's and Gaeta's group), was the excitation of threshold *instabilities and self-sustained oscillations in nonlinear counter-propagating waves* due to such a universal factor as regular linear dispersion. All these results have been also used and cited by many research groups.

In 1982 and later work [46], this PI theoretically predicted the most fundamental bistable/hysteretic effect in a cyclotron resonance of a single electron due to slight relativistic change of its mass. This prediction has been later on verified experimentally by Dehmelt and his group

[47], who has later on received Nobel Prize for unique experiments on single-particle trapping, one of which was the experiment [47] on hysteretic cyclotron resonance of a single trapped electron.

This PI has then broaden his research to *the relativistic nonlinear optics with single particles* [48] to such new predictions as optical high-order sub-harmonic excitation, high-order cycloraman scattering of laser by a single electron, etc. Significant part of these effects have been later observed in experiments by Wineland's group (C. S. Weimer et al, [49]); these research results of this PI have been used and cited by great many researchers including some very recent results (e. g. very recently by P. M. Visser and G. Grynberg's group [49]). He has also applied this theory to the nonlinear cyclotron resonances in semiconductors [48].

Beginning from 1984, this PI and his collaborators (S. Datta and C. T. Law) [50] worked out a new idea of new *sources of X-ray emission via transition radiation by low-relativistic electron beams traversing a solid-state nanostructures*. The fundamental new feature proposed in the recent work [51] by this PI, C. T. Law and P. L. Shkolnikov, was the use of *atomic absorption edges* of the layer components for excitation of very narrow transition radiation emission. Based on that effect and using our results [51], two groups (in Europe, P. Henri et al., and Japan, K. Yajima et al) [52] have observed narrow resonant transition radiation lines in various nanostructures using K-shell transitions, while the Verhoeven's group [52] is in the final stages of preparation of their experiment.

Continuing his research line on nonlinear wave propagation, in particular solitons and self-trapping, this PI has first discovered the existence of so called *bistable solitons*, or in general *multi-stable solitons* in both temporal and spatial domains [53]. This work has been immediately expanded by him in collaboration with the Canadian group of Dr. R. Enns [53] into the exploration of vast new landscape of *robust, weakly-stable and strongly-unstable* solitary and solitary-like waves of nonlinear Schrödinger wave equations with arbitrary nonlinearities. This work had had a large following, with great many researchers using its results and citing it. By now there are a few experimental observations of these new solitons, some of them very recent.

Since semiconductors were becoming a very promising nonlinear optical materials, especially as far as nonlinear refractive index, this PI has contributed into their development at its very first stage; he was part of the Purdue group which had first experimentally discovered in 1986 [54] large *nonlinear excitonic absorption* resonances in  $(\text{Zn}, \text{Mn})\text{Se}$  super-lattices and  $\text{ZnSe}$  films. Later on [54] he has also contributed into measurement of spectral measurement of the nonlinear refractive index in  $\text{ZnSe}$  using the effect of self-bending of a laser beam, developed by him and his group earlier [33]. He was also a part of a small group of top experts who wrote an assessment review paper on nonlinear optical materials in 1987 [55].

Having a long-standing interest in all aspects of nonlinear wave propagation, in particular in all kinds of solitons (see e. g. his work on bistable solitons [53] above), this PI together with his student & collaborators G. A. Swartzlander and D. R. Andersen, have predicted the existence of *spatial dark solitons* in the materials with negative nonlinear refractive index, and experimentally discovered and explored them [56]. The work [56] had had extensive following, with a few hundred citation by other workers; the research in the field is still current.

Being always interested in "behind the horizon" areas of research, this PI and his group, most of all Dr. P. L. Shkolnikov, were first to initiate in 1991 and extensively develop a new area

of quantum electronics, *X-ray nonlinear optics* [57-59], with this development driven by the need to have, in addition of very limited number of X-ray laser lines, as many sources & frequencies of coherent radiation in the X-ray domain, as possible. It included first research on X-ray resonant nonlinear effects in plasmas, search for "discharge plasma - X-ray lasers" resonant couples for X-ray nonlinear optics [57], X-ray harmonic generation in plasmas of alkali-like ions, X-ray laser frequency near-doubling (in collaboration with P. L. Hagelstein of MIT) [57], etc. This PI group also developed an extensive theory of *phase matching for large-scale frequency up-conversion* in gases and plasmas and other related problems [58]; and together with Drs. P. L. Shkolnikov and E. Hudis, this PI predicted a large *X-ray stimulated electronic Raman scattering* in *Li* and *He*, and *ionization-front solitons* in the X-ray stimulated Raman scattering, [59]. This work has been used by a many groups, including the first experimental observations of these effects.

Starting with his first results on two-level systems strongly driven by a laser with *arbitrary amplitude- and frequency-modulation* [60] in 1974 (see also [37]), in which he was first to develop an exact analytical theory of two-level system dynamics of very broad classes of laser modulation, which was later on used and cited by many workers in the field, this PI has always been interested in the laser interaction with these most basic objects of quantum electronics. Because in the recent discovery of the generation of very-high order harmonics in rare gases, there was a great deal of experimental and theoretical work on the nature of this phenomenon. This PI together with Dr. P. L. Shkolnikov in 1994 [61] put forward a hypothesis that this phenomenon might be explained by the hyper-polarization of a two-level system formed by the ground state of a rare gas and its far-removed first excited level (or a tight group of the excited levels), super-driven by a powerful laser, so that Rabi-splitting of the system far exceeds the energy of the two-level system at rest (they called it *a super-dressed two-level atom*). They developed a detailed theory of such an over-driven two-level system, and compared its results with huge amount of experimental data, and showed that this hypothesis can successfully compete with the Corkum's theory of ionized electron. The work [61] has been cited and used by many workers in the field, and the super-dressed atom is by now one of few "legitimate" models left in the field.

Since the effect of the so called cascade generation of second-order harmonics in  $\chi^{(2)}$  materials resulting in the emulation of  $\chi^{(3)}$  nonlinear refractive index, became of great interest as of recently, this PI has developed in 1993 the most basic theory of this effect by constructing the so called *eigenmodes of  $\chi^{(2)}$  wave-mixing* [62], and showed that they are a quint-essence of the cross-induced 2-nd order nonlinear refraction. This almost back-of-the-envelope model quickly became a popular tool in the field and has by now been used and cited by many researchers.

The presence of the top School of Medicine at the Johns Hopkins University was greatly encouraging and instrumental in initiating bio- and medical-related nonlinear-optics research of this PI's group. In collaboration with Dr. M. D. Stern, then a professor of cardiology at the JHU School of Medicine, this PI and his group conducted one of the first theoretical and experimental studies beginning at 1995 of *two-photon induced fluorescence of biological markers using optical fibers* [63]. They were the first to realize the potential of the technique for exploration and diagnostics of turbid and opaque bio-liquids such as e. g. blood in bulk instead of just its surface. They were also first to propose and observe the scheme [63], 1997, whereby the same optical

fiber was used to irradiate the sample and collect the scattered fluorescence; an amazing results was that the best performance was shown by a single-mode fiber. Albeit this research was later on discontinued because of lack of new grad-students experimentalists, its results have been used very recently by other researchers in developing new bio- and medical technologies based on two-photon induced fluorescence [64].

Most recently, in 1997 this PI in collaboration with Dr. P. L. Shkolnikov [73] and later on with Dr. J. Meyer-ter-Vehn group (Germany), have proposed to use available powerful lasers for *mass-production of elementary particles and nuclear reactions initiation* via *cascade* process. The idea was to (1) use the observed super-hot electron born when a sub-terrawatt femtosecond laser hits the surface of solid-state, with their energies being in the relativistic domain up to tens of *MeV*, to hit the other solid-state level of appropriately chosen material, (2) to excite the mass-production bremsstrahlung photons having roughly the same (or slightly lower) energies as the electrons; (3) these photons then while propagating in the next specifically chosen layer, where the cross-section of their interactions with nuclei of that material is orders of magnitude higher than that for the initial electrons, would initiate the nuclear reactions that should result in the elementary particle (such as electron-positron pairs, neutrons, protons, etc) production as well as other particle clusters and the internal excitation of nuclei. The calculations have shown potentials for the production of e. g.  $10^6$  neutrons per laser shot. This kind of processes can be developed into a practical source of ultrashort-pulse high-flux "nuclear radiation" (a shorthand for positrons, gamma-photons, neutrons, and fission fragments), with important applications to material science, medicine, and nuclear technology, such as gamma or neutron field interrogation and monitoring of radioactive waste, and the estimation of the effect of "full-scale" nuclear explosions on materials. In spite of its very recent publication, the results of work [73] has already been used by a few groups.

This PI has been a member of program committees and a panel member of several technical conferences on nonlinear optics and quantum electronics, and a member of PR committee of OSA. He is an editorial board member of the "International Journal of Nonlinear Optical Physics and Materials" (continuously) and "Optics Communications" (till the end of 1999). In 1996, this PI was awarded a prestigious Alexander von Humboldt Research Award for Senior US Scientists by the Alexander von Humboldt Foundation of Germany.

## **2. Final technical report on the AFOSR grant #F49620-99-1-0096 (E51-2048)**

Since this AFOSR grant started on January 1, 1999, a number of new results were obtained by this PI and his group in the field of nonlinear optics and quantum electronics, in the following directions:

- 2.i. Continuing pioneering research on sub-femtosecond pulses and physics with them, in particular  $\delta$ -ionization by super-short pulses and discovery of so called area resonances in ionization of quantum wells by sub-cycle field pulses
- 2.ii. Pilot pioneering research on nuclear gamma optics, in particular three-photon photon-nuclear fission
- 2.iii. Continuing pioneering collaborative research on fundamental new phenomenon of multi-mode interference in quantum mechanics, in particular highly regular pattern formation by



$\delta$ -excited wave functions.

- 2.iv. Discovery and study of novel effect of field-gradient-induced second-harmonic generation in nonlinear optics of vacuum.
- 2.v. Preliminary research on new perspective directions

### 2.i. Sub-femtosecond pulses and physics with them

Sub-cycle (unipolar, non-oscillating, half-cycle) electromagnetic pulses of *picosecond* and *sub-picosecond* duration have been available experimentally for more than a decade, see e. g. [65] and references therein. Recently, this PI and his group have theoretically shown the feasibility of even shorter, few- and sub-femtosecond (down to  $10^{-16}$  s) sub-cycle pulses and solitons of high intensity [26,27]. They were first to predict generation of high-intensity sub-femtosecond pulses by the multi-cascade stimulated Raman scattering [26] and by the so called EM-bubbles by a super-driven 2-level systems [27]; he also studied 3D propagation effects with these pulses [28]. We will call here such sub-cycle pulses "superpulses" (SP).

It is obvious that the next step of fundamental importance is to find out what is new physics due to these new pulses, in particular what are new features of their interaction with matter, especially in the situation where the pulse duration,  $\tau_{pl}$ , is shorter than a specific time of the major process, e. g. when it is shorter than the inverse frequency of a major quantum transition,  $\omega$ , so that  $\tau_{pl} < \omega^{-1}$  or even shorter than the *ionization* time of the system,

$$\tau_{pl} < \hbar/E_{ion} \quad (2.i.1)$$

In our research under this grant we considered two major new phenomena: the ionization of quantum wells and *H*-atom by superpulses (which we will call  $\delta$ -ionization), and "*area resonances*" in ionization of quantum wells.

The interaction of such pulses with quantum systems poses a substantial challenge to theory. Indeed, the slow-varying envelope approximation, a powerful tool of atomic and laser physics and nonlinear optics, does not make sense for superpulses; they are neither slow nor envelopes. This fact also translates into inapplicability of rotation-wave approximation due to the non-oscillating nature of the pulses. Another major factor is that the superpulses are too strong for the applicability of perturbation theory.

Therefore, one must use, or develop, techniques that do not depend on either slow-varying envelope, rotation-wave approximation, or perturbation theory. One such tool is an extension of impulse approximation, well known in the quantum theory of collisions, but has rarely been used in the theory of light-matter interactions [66]. To distinguish our (superpulse) version of impulse approximation, we will call it  $\delta$ -*model*; we will be talking, e. g., about  $\delta$ -ionization and  $\delta$ -excitation of atomic systems.  $\delta$ -model is valid when the pulse is much shorter than the evolution time of the respective quantum system, see e. g. (2.i.1). Then, the electric field of the pulse can be modeled by a Dirac delta-function of time. In the  $\delta$ -model, the effect of a SP on a quantum system is described by simply multiplying the pre-pulse wavefunction of the system by a coordinate-dependent phase factor. Namely, the wavefunctions just before ( $\psi_0$ ) and after ( $\Psi$ ) the pulse are related as follows:

$$\Psi = \exp[-(ieQ/\hbar)\sum r_j] \psi_0, \quad (2.i.2)$$

where the sum is taken over all the charged particles in the system [13], and *Q* is the *pulse area*

$$Q \equiv \int dt' \mathcal{E}(t') \quad (2.i.3)$$

with  $\mathcal{E}(t)$  being the pulse's electric field. The average momentum of the system changes then by  $\Delta \mathbf{p} = e\mathbf{Q}$ , while the change in the average *total* energy looks as a classical relation between the changes in the momentum and *kinetic* energy. For instance, for a one-particle system,

$$\Delta E \equiv \int d\mathbf{r} \Psi^* H_0 \Psi - \int d\mathbf{r} \psi_0^* H_0 \psi_0 = (\Delta \mathbf{p})^2 / 2m + \mathbf{p}_0 \Delta \mathbf{p} / m \quad (2.i.4)$$

where  $\mathbf{p}_0$  is the average momentum of the system in the initial state, and  $H_0$  is the system's Hamiltonian before the sub-cycle pulse (field-free Hamiltonian). Apparently, the pulse amplitude and duration are no longer meaningful if taken separately. Instead, the pulse's effect on the system is solely determined by  $Q$ . (By contrast,  $Q \approx 0$  for an oscillating pulse.)

### 2.i.1. $\delta$ -ionization by super-short pulses [1]

In our research under this grant, we have discovered [1] theoretically that ionization of a one-dimensional quantum well by an ultrashort sub-cycle ( $\delta$ -like) electromagnetic pulse results in photoelectrons propagating both along the field and in the opposite direction in well-separated shells, whose velocities reflect quasi-resonances in the ionization continuum. For both quantum well and a three-dimensional hydrogen-like atom, a unipolar, i. e. strongly *asymmetrical*,  $\delta$ -ionization produces an approximately *symmetrical*, stratified photoelectron cloud.

An important distinction of our approach is our focus on the photoelectron wavefunction in the coordinate representation. This focus is productive because of the unique, "trans-spectral" coherence of the  $\delta$ -ionization, as opposite to the ionization by a white-noise signal of a long timewidth with equally broad spectrum. It is this coherency that maps the energy-level structure of a quantum system into the ordered spatio-temporal structure of the photoelectron cloud, rather than into a random superposition of electron bunches that would result from a white-noise ionization. While the ionization of Rydberg atoms by a  $\delta$ -like electrical field "kick" has been considered before [66], in particular when a spatially-uniform  $\delta$ -function of time was used to approximate the field of a fast incident electron, our research was first to predict the spatio-temporal behavior of photoelectrons. We have investigated in more detail two fundamental quantum systems:

- \* *a 1D quantum well and*
- \* *a 3D hydrogen-like atom.*

Much of the symmetry of the  $\delta$ -ionization, however, is not related to particular potentials. It is easy to show, e. g., that for any real and inversion-symmetrical field-free Hamiltonian immediately after a highly asymmetrical (unidirectional)  $\delta$ -kick, the spatial distribution of photoelectrons is exactly symmetrical with regard to the field direction. Furthermore, the photoelectron *propagation* is approximately symmetrical with regard to the field direction. The symmetry is a purely quantum effect. Indeed, a classical particle hit by the unidirectional field of a sufficiently strong  $\delta$ -kick would move in the direction of the force. Quantum  $\delta$ -ionization, on the contrary, is similar to hitting water in a shallow bucket: a kick would spill water all over the rim (unless the kick is very strong, in which case almost all the water will move along the kick). (Likewise, the scattering of a particle off a power center is isotropic for small incident momenta.) This symmetry

holds for any inversion-symmetrical Hamiltonian.

For weak kicks, the velocities of the electron bunches (shells) correspond approximately to maxima in the transition dipole matrix elements. In other words, weak  $\delta$ -kicks simply map the structure of the photoionization dipole momentum into the photoelectron energy spectrum. As the pulse area grows, the spectral maxima and minima shift to higher (lower) energies for forward (backward)-moving electrons -- the effect that could be viewed as an SP analog of the Stark shift. Physically, this means that the velocities of the photoelectron bunches are determined, for a given quantum well, by the pulse area and might be used to measure areas of ultrashort pulses.

Investigation of  $\delta$ -ionization of hydrogen-like atoms has been greatly facilitated by the formal similarity of the amplitudes of the after-pulse wavefunction and amplitudes required by Born approximation, since the latter have been extensively researched, see e. g. [68]. The photoelectron spatial distribution shows apparent similarity with that for 1D quantum wells: an approximate symmetry with regard to the field direction and pronounced maxima (shells). Now, however, a shell does not necessarily correspond to electrons with close velocities, as is the case for a quantum well. Moreover, the spectra do not contain any peaks for weak kicks. We believe that the shells in hydrogen photoelectron cloud are in fact related to the 3D nature of the process and reflect strong spatio-temporal diffraction of the after-kick photoelectron cloud, which may result in the time-derivative behavior similar to that of initially nonoscillating EM pulses [28].

The effects considered here for hydrogen-like atoms could be observed in the ionization of Rydberg atoms, with  $t_{ion} \equiv E_{ion}/\hbar$  ( $E_{ion}$  being the ionization potential) in picoseconds, by existing sub-picosecond HCP, using e. g. the visualization technique of Ref. [69] in combination with time gating. Similarly, relatively long pulses can emulate a superpulse for the ground-state ionization of a shallow quantum well. Moreover, our simulations for *finite-width* superpulse ionization show many features of the  $\delta$ -ionization even for the pulses as long as  $\hbar/U_0$ . Therefore, with e. g.  $U_0 = 10 \text{ meV}$ ,  $\sim 200 \text{ fs}$  pulses will be short enough to observe the characteristic stratification; this duration is close to that of already available HCPs. An almost  $\delta$ -ionization-like picture would require about four times shorter pulses. The evolving stratified photoelectron distribution might be observed using a combination of near-field microscopy with ultrafast time-resolved spectroscopy proposed recently for studying other spatio-temporally localized electronic phenomena in semiconductor quantum wells [70].

### 2.i.2. Area resonances in excitation of quantum wells by sub-cycle field pulses

In our research [6,2] under this grant we showed theoretically that non-oscillating (sub-cycle) electromagnetic pulses much shorter than the evolution times of a quantum system may cause, aside from a  $\delta$ -ionization, resonant (preferential) transfer of population to a chosen quantum state for particular values of the pulse area. These "area resonances", which differ qualitatively from the presently known frequency resonances, hold a unique potential for control of chemical processes and solid-state devices.

Excitation and ionization of a quantum system by a  $\delta$ -like pulse field (an approximation of an ultrashort pulse with non-zero area) exhibit new kinds of resonances, both in the coordinate space (such as an ordered spatial stratification of the photoelectron cloud, see Section 2.i.1

above) and in the energy space (resonant population transfer between bound states, with related oscillations in the spread of the population over the continuum states). For resonances in the coordinate space, see [1].

In the energy space, the resonant population transfer from the ground state to the  $j$ -th occurs when the momentum  $p_\delta$  transferred from the pulse to the system,

$$p_\delta = e \int \mathcal{E}(t) dt = e Q \quad (2.i.5)$$

where  $Q = \int \mathcal{E}(t) dt$  is the pulse area as defined in (2.i.3) in the previous Section, is approximately equal to the *classical* quasi-momentum  $p_{cl}$  defined for the  $j$ -th state as:

$$p_{cl} \equiv \sqrt{2mT}, \quad T \equiv E_j - U, \quad E_j < 0 \quad (2.i.6)$$

In a quantum well, in particular,  $T = E_j + U_0$ . (Note that in quantum mechanics, the energy eigenstates do not possess a definite momentum, while the average momentum in bound eigenstates of definite parity is equal to zero.)

Our calculations [6,2] showed that the interaction of a quantum well with an ultrashort super-pulse exhibits resonances in the level population and in the photoelectron spectra for particular values of the pulse area. Qualitatively similar resonances exist in the interaction of super-pulses with hydrogen atoms. The effect is quite surprising, since an ultra-broad-band SP seems highly unlikely to resonantly excite a particular quantum transition. Moreover, since the spectrum of the  $\delta$ -pulse is flat, one would expect the population of the excited levels to mirror the distribution of dipole moment matrix elements, especially given that this is true in the perturbation limit. Indeed, for a perturbation that disappears at  $t \rightarrow \pm\infty$  [67], the amplitude of a  $j$ -th state is proportional to the dipole momentum spectrum:

$$a_j \sim d_{0j} \int_{-\infty}^{\infty} e^{i\omega t} \mathcal{E}(t) dt = d_{0j} \mathcal{E}_0 \quad (2.i.7)$$

This does not, however, hold at higher amplitudes; for sufficiently high  $p$ ,  $\delta$ -like interactions cannot be considered in the perturbation approximation at all. Instead, excitation by a  $\delta$ -kick is *resonant-like* : only levels separated from the ground energy level by approximately classical energy transfer are substantially populated. As we showed [6] for a number of fundamental quantum systems, conventional frequency resonances are replaced for SP by *area resonances* , which occur when the pulse area assumes specific values related to the electron eigenenergies. In a 1D quantum well, for example, the resonant population transfer from the ground state to the  $j$ -th state occurs when

$$p_\delta \approx p_{cl}, \quad p_\delta = e \mathcal{E}_p \tau_p = \int_{-\infty}^{\infty} e \mathcal{E}(t) dt, \quad p_{cl}^j \equiv \sqrt{2mT_j}, \quad T_j \equiv E_j - U, \quad E_j < 0 \quad (2.i.8)$$

where  $p_\delta$  is the classical (equal to the quantum average) momentum transferred to the system by the  $\delta$ -like pulse, and  $p_{cl}^j$  is the momentum of a *classical* electron that moves inside the well with the energy equal to the eigenenergy  $E_j$ . In a simpler case of an electron in an infinite quantum well, initially in the ground state, area resonances occur at

$$q = n\pi/2, \quad \text{with } n = 1, 2, 3, \dots; \quad \text{where } q \equiv p_\delta (2L)/\hbar, \quad (2.i.9)$$

with  $q = Q/e$  being a dimensionless pulse area, (2.i.3), when the population is excited mostly to the levels with  $j = n, n \pm 1$ . Moreover, at even  $n$  in Eq. (2.i.9) -- a momentum resonance to a



level, transitions to which from the initial state are allowed in the dipole approximation -- the ground state would become completely empty (except for  $q = \pi$ ).

This effect can be illustrated with the simplest case of the one-dimensional quantum box (QB) with infinite walls, with its potential being zero at  $0 \leq x \leq 2L$  and infinite otherwise. The eigenenergies of QB are

$$E_j^{QB} = (\pi j/2)^2 (\hbar^2/2mL^2) \quad (2.i.10)$$

and the eigenfunctions are  $\psi_j(x) = L^{1/2} \sin(j\pi x/2L)$ . One may define a quasi-momentum  $p_j$ , in such a way that  $E_j$ , Eq. (2.i.10), will have a classical appearance:

$$E_j = p_j^2/2m, \quad p_j \equiv (\pi j/2)(\hbar/L) \quad (2.i.11)$$

If before the  $\delta$ -kick the electron in QB is in the ground state,  $j=1$ , then after the kick its wavefunction is

$$\Psi(0+, x) = \exp(ip_\delta x/\hbar) \psi_1 \quad (2.i.12)$$

The population amplitudes after the kick, defined as  $\int \psi_j^* \Psi(0+, x) dx$ , are then as:

$$b_j = q F_j(q) \left[ \frac{1}{q^2 - [\pi(j+1)/2]^2} - \frac{(-1)^j}{q^2 - [\pi(j-1)/2]^2} \right], \quad (2.i.13)$$

where  $F_j(q) = -\sin(q)$  for  $j=3, 5, \dots$  and  $F_j = i \cos(q)$  for  $j=2, 4, \dots$ , whereas for the ground state,  $j=1$  we have:

$$b_1 = \frac{\sin(q)}{q} + \frac{1}{2} \left[ \frac{\sin(\pi+q)}{\pi+q} + \frac{\sin(\pi-q)}{\pi-q} \right] \quad (2.i.14)$$

We can see from Eq. (2.i.13) that we have an area-resonant excitation. i. e. maxima of  $|b_j|^2$  occur, for a given  $q$ , if

$$q = \pi n/2, \text{ or } 2q/\pi \text{ is an integer } n \quad (2.i.15)$$

For such "quantized" momentum transfer, the maxima in the spectrum  $b_j$  are particularly sharp: half of all the transferred population concentrates in just two levels with  $j = n \pm 1$ . This resonant condition may also be expressed as a "quasi-momentum resonance": if the momentum transfer is equal to the quasi-momentum of one of the QB eigenstates,

$$p_\delta = p_n, \quad (2.i.16)$$

one observes sharp concentration of population at the levels adjacent to the  $n$ -th level, with the maximum on the  $n$ -th level itself. The respective amplitudes are  $b_{n \pm 1} = 0.5$  and  $b_n \approx 2/\pi$ , so that these three levels will contain  $\approx 0.9$  of all the population.

Particularly interesting is the case of even  $n$  in Eq. (2.i.15),  $n \equiv 2l$ ,

$$q = l\pi \quad (2.i.17)$$

In other word, we consider a momentum resonance to a level transitions to which from the initial (ground) state are allowed in the dipole approximation. Such pulses would completely empty the ground state. Let us calculate the "survival probability" -- the probability (2.i.14) that the electron will remain in the ground state after the kick. This amplitude zeroes out at  $q = n\pi = 2m(\pi/2)$ ,  $n$  integer -- i. e. exactly Eq. (2.i.17). (The only exception is  $q = \pi$ .) Therefore,

momentum resonances to the dipole-allowed transitions, except for the lowest area resonance, are accompanied by the total removal of the population from the ground level. A closer look at  $b_j$  reveals that, in fact, if  $q = n\pi$ , *all* levels with odd numbers are empty, except for  $j = m + 1$ . In particular, the weakest pulse with the momentum resonance to a dipole-allowed transition,  $m = 2$ , does not empty the ground level. On the other hand, for large  $q$ , the ground level population is small anyway, so that the resonant removal of population from the ground state is pronounced only for  $m = 4$ , or  $q = 2\pi$ .

In a finite quantum well, similarly, super-pulses that are area-resonant to the even-number levels, remove all the population from the ground (initial) state. Moreover, we have shown that a new surprising effect occurs in the ionization of this system: at the "dipole-allowed" area resonances, the spectrum of ionized electrons sharply concentrates near the bottom of the continuum, while at the area resonances to the odd-number bound levels, the photoelectron spectra are maximally broad.

## 2.ii. Nuclear gamma optics: laser-based multiphoton-photon photon-nuclear fission

Recent theoretical and experimental study of plasma driven by high-intensity ultrafast lasers have demonstrated generation of substantial amounts of relativistic electrons with energies in MeV range [71,72]. Based on some of those results, we showed theoretically [73] under the previous AFOSR grant that already attainable laser intensities are sufficient for producing, through a specially arranged cascade of processes, practically useful ultra-short-pulse, high-flux nuclear radiation ( $\gamma$ -photons, positrons, and neutrons) with possible applications in material science, medicine, and nuclear engineering. Moreover, even widely available table-top lasers are capable of generating, through relativistic laser-plasma electrons interacting with optimal targets, noticeable amounts of nuclear radiation and could, therefore, be instrumental in proof-of-principle experiments for future practical laser-based sources of that radiation.

In our research under this grant [10], we explored the ramifications of our research [73] and the largely unnoticed fact: the fluxes of relativistic electrons attainable in sub- and over-critical plasmas driven by sub-picosecond terawatt lasers exceed by many orders of magnitude the electron fluxes available at several MeV energy electron accelerators. Indeed, the highest electron current density available at electron accelerators for a few-MeV electron energy is about  $j \sim 10^7 \text{ A/cm}^2$ . At the same time, recent computer simulations [71] predict electron fluxes many orders of magnitude higher:  $j \sim 10^{11} \text{ A/cm}^2$  of few-MeV electrons in subcritical plasma at laser intensities between  $10^{19}$  and  $10^{20} \text{ W/cm}^2$ , and up to  $10^{13} \text{ A/cm}^2$  in solid-density plasmas. (The latter number corresponds to the Alfvén limit of the current,  $I_{\text{Alf}} = mc^3 \gamma e$ , which, however, might be exceeded locally by up to three orders of magnitude if sufficiently large return current is present.) These simulations are apparently corroborated by experimental results obtained at the Petawatt laser at LLNL [72]. As a consequence, which we propose to exploit experimentally, those electrons may generate, in optimal targets,  $\gamma$ -photons of sub-MeV and few-MeV energy with *unique intensity*. In fact, experimental data from a new Petawatt laser at LLNL apparently show  $\gamma$ -photons with  $\hbar\omega_\gamma \geq 2 \text{ MeV}$  generated in a highly-directional,  $\sim 5 \text{ ps}$ -long and  $\sim 30 \text{ }\mu\text{m}$ -wide "beam" with the total energy that reaches at least 0.5 J even for a far from optimal target (these numbers are inferred from the results Ref. [72]). This yields  $> 10^{16} \text{ W/cm}^2$  intensity of the  $\gamma$ -radiation -- of the same order of magnitude as the intensity available from focused high-intensity optical lasers. We estimate that target optimization, as well as

refinement of the laser beam, may increase this number by two orders of magnitude.

Another distinct advantage of these "laser-target" sources is their ability to combine the source and the target. Indeed, relativistic electrons and  $\gamma$ -photons would be generated in the material whose interaction with electrons or  $\gamma$ -photons is an application; this would eliminate the losses inevitably associated with transporting radiation from a source to a target. For instance, electro- or photonuclear fission may be studied in uranium which, as a high-Z material, is highly efficient for generating  $\gamma$ -photons via Bremsstrahlung.

We expect that the  $\gamma$ -field intensities and fluxes of MeV electrons from the proposed laser-target sources would be so high that the new regime in photo- and electronuclear physics might become feasible; we call this regime *nonlinear photonuclear physics*. To evaluate the feasibility of nonlinear photonuclear effects, we take into account the broad spectra of laser-target sources, which at this point limits our consideration to those nuclear effects for which monoenergetic radiation is not necessary, while very high intensity is. Fortunately, the most pronounced feature of the photonuclear absorption, nuclear Giant Dipole Resonance (GDR), which contains most of the photo-absorption oscillator strength and whose energy in actinides is close to the energy of a substantial fraction of relativistic laser-plasma electrons, is extremely broad ( $\Delta E/E \sim 0.3$ ). For actinides, such as uranium (U) and thorium (Th), the peak of the GDR cross section is at  $\sim 10\text{--}12\text{ MeV}$ , while the photo-fission threshold is just below  $\sim 6\text{ MeV}$ . Our estimates show that the proposed by us laser-based particle sources would be intense enough to observe the most fundamental nonlinear photonuclear process -- *triggering nuclear fission by multi- $\gamma$ -photon excitation* of the nuclear giant dipole resonance in U and Th.

The second kind of proposed by us high-flux sources of sub- and few-MeV  $\gamma$ -photons, which we will call "laser-beam" sources, is essentially an extension of laser-synchrotron sources, whose ability to generate high fluxes of hard X-rays (in tens of KeV) by scattering of optical laser radiation off accelerated electron beams is currently under active investigation, see e. g. [74]. If one replaces the beam of accelerated electrons with relativistic ultra-dense electron currents in plasma to backscatter short-wavelength laser photons, one may obtain an intense source of sub-MeV  $\gamma$ -radiation (with a soft X-ray laser), or a TW source of few-KeV hard X-rays (with an excimer laser). Moreover, in combination with high-energy ion accelerators, soft-XRL with the photon energy around  $300\text{ eV}$  (for the lasing in the "water window", which is rapidly becoming available, may provide yet another source for nonlinear photonuclear physics. Indeed, Large Hadron Collider, to be in operation around 2005, will accelerate heavy ions to the relativistic factor of  $\sim 3000$ . Such ions would "see" the Doppler-shifted photon energy of a  $300\text{ eV}$  XRL as  $1.8\text{ MeV}$   $\gamma$ -radiation; the field intensity in the rest system of an ion would be  $4\gamma^2 \sim 4 \times 10^7$  larger than that in the XRL's rest system and may exceed  $10^{23}\text{ W/cm}^2$  for the XRL output of a few MW.

Another unique application of laser-based  $\gamma$ -sources to nuclear physics may be generating fast-fissioning (on the nanosecond time scale) shape isomers in substantial amounts. These isomers exist for fissioning nuclei with a double-humped fission barrier and correspond to the states located in the more shallow well, 2-3 MeV above the main ground state. The new sources may also be used for intense broad-band excitation of nuclei in nuclear spectroscopy.

In QED, using a crude version of a  $\gamma\text{--}\gamma$  collider ( $\gamma$ -radiation from two targets irradiated by multi-TW laser beams, which might yield up to a 1000 events per pulse) or scattering of X-ray

laser radiation off a GeV electron beam (e. g., Y X-ray laser and 6 GeV beam, with a few events per pulse), one may observe, for the first time, the Breit-Wheeler reaction  $\omega_1 + \omega_2 \rightarrow e^+ e^-$ .

Among practical applications of high-intensity, ultrashort-pulse laser-based  $\gamma$  and hard-X-ray sources, we suggest: testing the behavior of materials under the conditions similar to a nuclear explosion, by subjecting these materials to high-intensity pulses of nuclear radiation generated using lasers; high-energy flash radiography and cineradiography for studying explosions and other fast changes in bulk solids; soft- and hard-X-rays for medical imaging, material science and processing; and developing positron and neutron sources with high temporal and spatial resolution and very low radiation hazard.

### 2.iii. Multi-mode interference: Highly regular pattern formation in quantum mechanics

In the research under the previous AFOSR grant started by this PI [75] in collaboration with Dr. W. P. Schleich and his group and Prof. W. E. Lamb, Jr, it was first demonstrated that highly regular spatio-temporal or multi-dimensional patterns in the quantum mechanical probability or classical field intensity distributions can appear due to pair interference between individual eigen-modes of the system, thus forming the so called intermode traces. These patterns are strongly pronounced in any anharmonic potential, provided that the traces are multi-degenerate; they may occur in many areas of wave physics. These traces have been observed earlier in numerical simulations of the evolution of a Gaussian wave packet [76], an initially homogeneous wave function [77] or well localized wave packets [78], a Bose-Einstein condensate [79], and an angular wave packet [80]. The major contribution of this PI was to identify the multi-mode interference nature of the process and generalize it to arbitrary potentials. The work [2,4,7,13] under the reported grant is the continuation of that research.

The controlled preparation and measurement of wave packets has recently emerged as an extremely active field of research. Examples include wave packets in atoms [81], optical lattices [82], ion traps [83], molecules [84], and semiconductor quantum wells [85], to name a few. One of the major points of interest is the spatio-temporal evolution of wave packets in such systems. Since most of the experiments engage a broad-spectrum excitation of the eigen-modes of the system, large-scale interference becomes a leading factor. It gives rise to well-ordered long-range regularities such as quantum revivals [86], interference patterns in the atomic double-slit experiment and diffraction effects in atom optics [87] and  $\delta$ -ionization [1]. We showed [75,2,4] that these packets may also serve as a testing ground for a new class of interference effects: the formation of highly regular spatio-temporal or multi-dimensional patterns in the quantum mechanical probability,  $|\psi|^2$ , or classical electromagnetic field intensity,  $|\mathcal{E}|^2$ . In all cases, the probability distribution is characterized by a regular net of *canals*, minima of probability, and *ridges*, maxima of probability, which run along straight lines in space-time  $(x, t)$ .

Similar patterns also arise in the near field of a diffraction grating illuminated with light [77] as well as in matter waves in atom optics [87]. Indeed, the paraxial approximation allows for a space-time analogy between quantum mechanics and electrodynamics. The space-time probability distribution  $|\psi(x, t)|^2$  is mapped onto the field intensity  $|\mathcal{E}(x, z)|^2$ , where  $z$  is the propagation direction. In the simplest case of the square well (or plane geometry of EM-diffraction grating) this phenomenon reveals a new aspect of the so-called Talbot effect [88], which

traditionally concerns only patterns in planes at *fixed* time  $t$  or distance  $z$ , i. e. "slices" of the distribution, instead of the patterns in the "full propagation" space  $(x, t)$  or  $(x, z)$ .

The common feature of these systems is a broad-spectrum excitation: e. g. in the quantum case, the rich patterns appear only when many states are populated. So far the theory of these patterns has relied on the specific choice of the initial conditions and on the square well (or diffraction grating structure), with no simple physical explanation suggested. In our recent work [15] we observed similar structures in both the square well and a smooth potential and identified the key mechanisms of the phenomenon as pair multi-interference between the eigen-modes of the system and the degeneracy of this interference.

To illustrate the basic theory of intermode traces, we consider here only one-dimensional (1D) problems, that is problems which involve either spatio-temporal patterns in quantum mechanics with one coordinate and a time variable, or spatial patterns in optics with one transverse and one longitudinal variables. A similar phenomenon in higher dimensions will be considered in a similar way. To study the time evolution of a particle moving in a 1D binding potential  $U(x)$ , we represent the full wavefunction as as the superposition of "WKB" eigen-wavefunctions within the WKB approximation [89] as:

$$\psi_n^{(WKB)}(x, t) \approx f_n(x) \exp[\pm i \int k_n(x) dx - i E_n t / \hbar]. \quad (2.iii.1)$$

where both the pre-exponential factor  $f_n(x)$  and the momentum

$$\hbar k_n(x) \approx \sqrt{2m_e[E_n - U(x)]} \quad (2.iii.2)$$

are assumed to vary slowly compared to  $\exp[\pm i \int k_n(x) dx]$ , and  $E_n$  are the eigen-energies of the system. We showed that the space-time trajectories which define the traces are determined by the pair of eigen-functions of the  $n$ -th and  $m$ -th order, and are essentially the lines of constant phases, so their phase velocities,  $v_{nm}(x)$  are as:

$$v_{nm}(x) = \left( \frac{dx}{dt} \right)_{nm} = \frac{(\Delta\omega)_{nm}}{(\Delta k)_{nm}}, \quad (2.iii.3)$$

where  $(\Delta\omega)_{nm} \equiv (E_n - E_m)/\hbar$ , and  $(\Delta k)_{nm} \equiv \pm [k_n(x) \pm k_m(x)]$ , or

$$v_{nm}(x) \equiv \pm \frac{\omega_n - \omega_m}{k_n \pm k_m} \approx \pm \frac{(E_n - E_m)/\sqrt{2m_e}}{\sqrt{E_n - U(x)} \pm \sqrt{E_m - U(x)}} \quad (2.iii.4)$$

where  $\omega_n \equiv E_n/\hbar$ . The trajectory is then  $t = \int_{x_I}^x v_{nm}^{-1} dx$ , where  $x_I$  is the turning point for the lower energy  $E_n$  and  $E_m$ . For every pair of quantum numbers  $n$  and  $m$  we find *four* traces, with all the possible combination of signs in (2.iii.4).

When  $\Delta\omega, \Delta k$  are large, the velocity  $v_{nm}$  approaches the phase velocity, and describes strictly quantum features of the motion. In contrast, when  $\Delta\omega, \Delta k$  are small,  $v_{nm}$  is reminiscent of a group velocity corresponding to a classical motion of a particle. Indeed, an initially almost classical motion is described by a compact group of eigen-modes near some high quantum number  $N$ , with a number  $\Delta N$  of these modes satisfying the condition  $1 \ll \Delta N \ll N$ . This excitation results in a strong *clustering of traces in two groups*. The one with

$$|(\Delta\omega)_{n,m}| \ll \omega_N, \quad \text{and} \quad (\Delta k)_{n,m} \ll |k_N|, \quad (2.iii.5)$$

is essentially a *classical* trajectory, since in such a case

$$\frac{(\Delta\omega)_{n,m}}{(\Delta k)_{n,m}} \approx \frac{d\omega}{dk} \equiv v_{gr}, \quad (2.iii.6)$$

where  $v_{gr}$  is a group velocity. It coincides with the classical velocity, since the intermode trace equation obtained from (2.iii.4) as

$$\frac{dx}{dt} = \sqrt{2[E_N - U(x)]/m_e}, \quad (2.iii.7)$$

describes classical motion of a particle with energy  $E_N$  in a potential  $U(x)$ . One can see now that the other group of traces, with

$$v_{nm} \ll v_{gr}, \quad \text{and} \quad |k_n \pm k_m| \approx |2k_N|, \quad (2.iii.8)$$

reflects the *quantum* behavior. Remarkably, the WKB approximation developed basically as a tool to describe quasi-classical motion, can also serve to describe highly quantum objects like the "quantum" group of intermode traces. The full set of velocities  $v_{nm}$  (2.iii.4), ranging from group velocity to phase velocity at the extremes, provides a greatly useful new tool in the understanding of quantum system.

As an illustration, we consider first the simplest potential, the square well potential with infinite walls. In the EM analogy, this corresponds to an EM wave in a waveguide with ideal metallic walls. In this case, the WKB results become *exact*, so that for a box of width  $L$ , the expression for the trace velocity (2.iii.4) reads

$$v_{nm} = \pm M \cdot v_{\square}, \quad M = m \pm n; \quad (2.iii.9)$$

where  $M$  is the normalized velocity, and  $v_{\square} = \pi\hbar/2mL \ll c$  is a characteristic velocity of a box.

Since  $M$  in Eq. (2.iii.9) is an integer, a trace with a certain  $M$  is attributed to *all* the couples of modes  $n$  and  $m$  whose sum or difference is  $M$ . If the number of the states involved is sufficiently large, this trace degeneracy creates multiple superimposed traces. They in turn give rise to the distinct straight canals. We have observed in computer simulations some of the traces predicted by Eq. (2.iii.9). Here we assume that the initial wave packet is the ground state wavefunction, i. e.  $\psi_I(x) \propto \sin(\pi x/L)$ . The system is hit by a  $\delta$ -pulse delivering a momentum  $\bar{p}$ , which we normalize to the ground state momentum  $p_I = \hbar k_I$ . We show the time evolution of  $|\psi|^2$  from  $t=0$  to  $T/2$ , with  $T \equiv 2\pi/\omega_I$  being the revival time.

When the excitation is relatively low,  $\bar{p}=1$ , only two eigenstates are involved. The classical trajectory bouncing between the walls, does not fit any pattern yet even though it has the same velocity as some of them. There are only two traces, not well fitting either: a "near-classical" trace created by selecting  $k_n - k_m$  in Eq. (2.iii.4) (or normalized velocity  $M = n + m$  in Eq. (2.iii.9)) with  $(n,m)=(1,2)$ , and a "quantum" trace, white dashed line, corresponding to  $k_n + k_m$  in Eq. (2.iii.4) (or  $M = n - m$  in Eq. (2.iii.9)).

For stronger excitation,  $\bar{p}=3$ , there are several traces with the same velocities fitting the straight patterns which emerge now more clearly. The black solid line for a near-classical trace is twofold degenerate since pairs (2,3) and (1,4) lead to the same  $M = n + m = 5$ . The quantum trace (broken line) is highly degenerate, being produced by pairs (1,2), (2,3), (3,4) and (4,5) with  $M = |n - m| = 1$ .

A more developed quantum traces have been simulated for  $\bar{p}=5$ . Beside the classical trajectory, and a near-classical trace for  $M = n + m = 9$  [with the pairs (3,6) and (4,5)], we observed



two examples of quantum intermode traces: for  $|n-m|=1$ , generated by pairs (3,4), (4,5), (5,6) and (6,7), and for  $|n-m|=2$  with pairs (3,5), (4,6) and (5,7).

It is obvious that the phenomenon is not restricted to the square well; it should occur for other potentials. Distinct patterns near *the classical trajectory* are better pronounced for strongly anharmonic potentials with "hard walls", the extreme example of which is a box, and less for "soft" potentials, e. g.  $U \propto |x|^w$  with  $w \leq 2$ , including the harmonic potential,  $w=2$ . This is explained by a high trace degeneracy in a box, whereby many individual traces with the *same* velocity bundle together to form the patterns; the soft potentials, on the other hand, create less degenerate traces. To illustrate this, we note from the definition of the trace velocity (2.iii.4) that the peak velocity, i. e. the velocity at  $U(x) = 0$ , is

We have developed the theory of the intermode traces in the se potentials in our work [2,4,75]. A particular example considered by us, was a potential which represents an intermediate case between the square box and harmonic potential. We have observed the quantum traces for the anharmonic potential

$$U = Ax^4, \quad (2.iii.10)$$

in which the initial wave packet is again a ground-state wavefunction hit by a  $\delta$ -pulse delivering momentum  $\bar{p}$ . Here we have introduced the system  $m_e = \hbar = A = 1$ , that is we have scaled  $x$ ,  $t$  and  $p$  by the factors  $x_s \equiv (\hbar^2/m_e A)^{1/6}$ ,  $t_s \equiv (m_e^2/\hbar A)^{1/3}$ , and  $p_s \equiv m_e x_s/t_s$  respectively.

We have simulated first the case of low excitation,  $\bar{p}=1$ , with only two states excited. Here, the classical trajectory (white solid line) does not produce a good fit yet while canals fit approximately to classical traces (black solid line), and ridges -- to quantum traces (white dashed line) with  $(m,n) = (1,2)$  in both cases.

We have further simulated the case of higher excitation,  $\bar{p}=3$ , with an almost classical distribution of state populations,  $\sigma_{nn}$ . Here the traces are well pronounced. The classical trajectory fits snugly into maxima of  $|\psi|^2$ . Moreover, now the near-classical trace with pair  $(m,n) = (2,3)$ , fits very well the canal patterns. The quantum trace for the same pair also fits the respective patterns.

Finally, when  $\bar{p}=5$ , we saw a pronounced classical motion for the first couple of cycles which then turns into a richly developed carpet well described by intermode traces. The dark solid lines are near-classical traces produced by the pair  $(n,m) = (5,6)$ , while broken lines denote a few examples of quantum traces: the dash-dotted one is created by the same pair, whereas the dashed one by (5,7).

To excite many atomic eigen-states using a *ground* state as an initial wave packet, instead of Rydberg states, one needs to shake-up the system by a strong  $\delta$ -like EM pulse shorter than a cycle of any eigen-frequency; potential avenues to generate these pulses are multi-cascade stimulated Raman scattering, molecule modulation, subfemtosecond field solitons, and very high harmonic generation [90]. However, in solid-state quantum wells [85] sub-picosecond half-cycle pulses can achieve the goal, while in atom optics of ion traps [83], with the motional frequency being in radio-domain, even a *nano*-second laser pulse can serve as a  $\delta$ -kick.

Insofar as the EM wave equation in paraxial approximation is isomorphous to the Schrödinger equation, the intermode traces can readily be found in optics and electrodynamics, with waveguides, resonators and spatially- or time-periodical structures providing a natural

playground for EM mode interference. For example, the modes of a sufficiently wide,  $L \gg \lambda$ , waveguide with ideally conducting walls (as well as the field scattered by a diffraction grating in the Talbot effect) have the same patterns as those of the square well, Eq. (2.iii.9). The modes of a regular fiber waveguide produce patterns similar to those of the box of finite width, while modes of a fiber waveguide with smoothly varying refractive index make patterns similar to those of a quantum well with the respective potential.

The intermode trace phenomenon can be extended to other areas of wave physics. Its existence does not depend on exact form or order of the wave equations; the linearity though is required to produce eigen-modes. One can relate well known wave phenomena such as the so called Kikuchi lines in X-ray diffraction in crystals [91], Chladni patterns in acoustics, and the formation of straight patches of calm surface in rough seas, to intermode traces. In more general terms, even nonlinear wave equations, such as e.g. nonlinear Schrödinger, Kordeweg-De Vries, and sine-Gordon, can support multi-soliton solutions, with the individual soliton trajectories reminiscent of intermode traces. The straight multi-solitons traces are e. g. found [27] in the modified Kordeweg-De Vries equation approximating the propagation of EM-bubbles. Highly organized two-dimensional nonlinear-optical patterns are formed both in the near- and far-field areas by the grid of spatial dark solitons [56], in a resonator filled with a Kerr-like nonlinear material [92], and by "scars" in a quantum billiard [93]. Similarly to intermode traces, these wave phenomena might be viewed as multi-wave effects resulting in well-organized "carpet" macro-structures in systems with a broad-spectrum excitation.

#### 2.iv. Nonlinear optics of vacuum: Field-gradient-induced second-harmonic generation

In his research [3] under this grant, this PI in collaboration with his former grad-student, Dr. Y. J. Ding have shown that photon-photon scattering in vacuum can give rise to the second-harmonic generation of intense laser radiation in a *dc* magnetic field in the "box" diagram approximation of quantum electrodynamics if the symmetry of interaction is broken by *modulation or/and nonuniformity* of optical wave + *dc* field system in time or/and space. Specific examples considered by them were: an optical *pulse* plane wave or a Gaussian laser beam propagating in uniform or nonuniform *dc* fields.

Photon-photon scattering (PPS) [94] is perhaps one of the most fundamental quantum electrodynamics (QED) processes which may also result in nonlinear optical effects in vacuum such as the birefringence of the refractive index seen by a probe field under the action of either a *dc* magnetic (or electric) field [95] or intense laser pumping [96], multiwave mixing [97], and merging of two photons into one (i.e. sum frequency generation) [98] under the action of a *dc* field. All of these effects are based on the lowest order, so called "box" diagram approximation, (It was also proposed that using hexagonal diagram, high-order harmonics [99] and second-order subharmonic, a so called photon splitting [95,100], can be generated with a *dc* magnetic field.) If observed, these effects may provide a fundamental optical test of QED.

From any realistic point of view the only hope to attain observable effects in the laboratory is to use lowest order processes (i.e. those due to the box diagram); yet the required optical fields are still enormously high and not presently available. It has also been apparent that a *dc* field (either electric or magnetic) may greatly assist the interaction, which have attracted much attention [95-100]. One of the most interesting and fundamental are *dc* field-assisted processes: the merging of two photons into one, e.g. the second harmonic generation, SHG



(see also [101]), and photon "splitting" [98,95,100,102], -- in essence, a parametric process -- in the presence of a  $dc$  field. However, these processes have been the subject of a long persisting controversy, with the work [98,95,100] maintaining (correctly) that these effects vanish in the box approximation (and under condition of  $cw$  wave and uniform and constant  $dc$  field, which is an important point in the context of this research), whereas the work [101,102] suggested (erroniously, see [95,100,103]) nonvanishing effect for the same approximation and conditions. The previous works on the subject have been reviewed in [103] with a conclusion that none of them were fully correct, although the results [103] are much closer to those of [98,95,100] (the results of [103] suggest the box diagram contribution for the plain wave is non-zero, yet smaller than that from the next order, hexagonal, diagram). The discussion [105] following proposal [104] of laser-induced SHG in vacuum in the presence of a  $dc$  magnetic field (see also earlier work [6]), pointed out that SHG for a  $cw$  plane wave should vanish. While agreeing with the major point of [105] that SHG due to the box diagram vanishes for a  $cw$  plane wave and uniform  $dc$  field (which was also the main case considered earlier [95-103]), the point of our [104] and later consideration [106] suggested, however, that the nonvanishing effect may result from the spatial *nonuniformity* of the field.

Hence, the fundamental question arises whether the vanishing contribution of the box diagram is due to fundamental laws of QED, or only due to the conventionally (and thus unfavorably) chosen configuration: plane monochromatic ( $cw$ ) optical wave + uniform  $dc$  field. In our recent work [3], we showed that the nonvanishing contribution of the box diagram can result from the *nonuniformity (or gradient) of any component of the entire field system* (both the laser and  $dc$  magnetic fields) in *space* or/and *time*. In particular, we considered plane wave modulated in *time* by a pulse with an arbitrary profile and finite duration, and a  $cw$  Gaussian (i. e. spatially *inhomogeneous*) laser beam in magnetic field with an arbitrary spatial distribution, in particular in both *uniform* and *nonuniform* fields -- magnetic dipole and quadrupole. We demonstrated that the nonvanishing SHG in the lowest (i.e. box) approximation exists in all these configurations. The most important fact is that the SHG effect (i) exists in the approximation in which the plane-wave SHG vanishes completely, and (ii) that it is of the expected order of magnitude; for example, for a Gaussian laser field with maximum amplitude,  $E_1$ , in the focal plane in a uniform  $dc$  magnetic field,  $B_0$ , the efficiency of SHG conversion,  $w_{SHG}$ , is

$$w_{SHG} = (\alpha/45)^2 O \left[ (E_1 B_0 / B_{cr}^2)^2 \right], \quad (2.iv.1)$$

where  $B_{cr} \equiv m_0^2 c^3 / e \hbar = 4.4 \times 10^9$  Tesla is the QED critical field, and  $\alpha = e^2 / \hbar c = 1/137$  is the fine structure constant. The-state-of-the-art of laser and magnet technology may make the observation of vacuum SHG feasible in near future. Our interpretation of field-gradient induced SHG in vacuum relates the field nonuniformity to momentum transfer between photons and  $dc$  field. The total number of SHG photons in such a system is many orders of magnitude higher than that due to the next, hexagonal diagram contribution, such that SHG appears to be truly of the "box diagram" nature.

In our first-principles-based calculations, we used the Heisenberg-Euler Lagrangian [94] for PPS  $L = L_2 + L_4 + L_6 + \dots$ , where  $L_2 = (E^2 - B^2)/2$  is a linear term, and

$$L_4 = (\chi/2) (a^2 + b^2); \quad (a \equiv E^2 - B^2; \quad b \equiv \vec{E} \cdot \vec{B}), \quad (2.iv.2)$$

is the first nonlinear term; it corresponds to the box Feynman diagram. Here

$\chi = \alpha/45\pi B_{cr}^2 = 2.6 \times 10^{-24} \text{ Tesla}^{-2}$  is a nonlinear interaction constant and  $B_{cr}$  is the QED critical field, see above.  $L_6$  corresponds to the hexagonal Feynman diagram, etc. Using the action for the first two terms,  $\int (L_2 + L_4) d^4x$ , and taking the variation with respect to the four-vector potential, the macroscopic equations in the form of classical Maxwell's equations are obtained as [94,96,108]:

$$\nabla \cdot \vec{B} = 0, \quad \nabla \cdot \vec{D} = 0; \quad \nabla \times \vec{E} + \partial \vec{B} / \partial t = 0, \quad \nabla \times \vec{H} - \partial \vec{D} / \partial t = 0, \quad (2.iv.3)$$

with the constitutive relations [94,108] between the electric displacement  $\vec{D}$  and magnetic field  $\vec{H}$ , and electric field  $\vec{E}$  and magnetic induction  $\vec{B}$  being:

$$\vec{D} = \vec{E} + \vec{D}^{NL}, \quad \vec{H} = \vec{B} - \vec{H}^{NL}, \quad (2.iv.4)$$

where

$$\vec{D}^{NL} = \partial L_4 / \partial \vec{E} = \chi (2a \vec{E} + 7b \vec{B}); \quad \vec{H}^{NL} = \partial L_4 / \partial \vec{B} = \chi (-2a \vec{B} + 7b \vec{E}). \quad (2.iv.5)$$

These equations are valid for  $|E|, |B| \ll B_{cr}$  and  $\lambda \gg \bar{\lambda}_C$ , [108,109], where  $\lambda$  is the field wavelengths and  $\bar{\lambda}_C = \hbar/m_0c = 3.862 \cdot 10^{-3} \text{ \AA}$  is the Compton wavelength of electron.

A *cw* plane wave does not exhibit any nonlinear effects, since due to its properties,  $E^2 = B^2$ ,  $\vec{E} \cdot \vec{B} = 0$  (i. e.  $a = b = 0$ ), the nonlinearity, Eq. (2.iv.5), vanishes. This "degeneracy" of the *third-order* nonlinearity may be broken by the field nonuniformity that can give rise to *second-order* nonlinear *optical* effects in the presence of a strong *static* field. If an unperturbed fundamental wave,  $\vec{E}_1$  &  $\vec{B}_1$ , propagates in vacuum in the presence of a *dc* magnetic field,  $\vec{B}_{dc}$ , Eq. (2.iv.5) can be rewritten as

$$\vec{D}^{NL} = \chi (-2B_{dc}^2 \vec{E}_1 + 7b_E \vec{B}_{dc}) + \vec{D}^{(2)}; \quad \vec{H}^{NL} = 2\chi B_{dc}^2 \vec{B}_{dc} + 2\chi (B_{dc}^2 \vec{B}_1 + 2b_B \vec{B}_{dc}) + \vec{H}^{(2)},$$

where  $b_B = \vec{B}_1 \cdot \vec{B}_{dc}$ ,  $b_E = \vec{E}_1 \cdot \vec{B}_{dc}$ , and the only terms that may give rise to SHG are

$$\vec{D}^{(2)} = \chi (4b_B \vec{E}_1 - 7b_E \vec{B}_1); \quad \vec{H}^{(2)} = \chi (4b_B \vec{B}_1 + 7b_E \vec{E}_1). \quad (2.iv.6)$$

Suppose now that the unperturbed fundamental light beam at the frequency  $\omega_1$  is a quasi-plane and/or quasi-*cw* wave with the wave-vector  $\vec{k}_1 = \vec{q}_1 k_1$ ,  $k_1 = \omega_1/c$ ,  $q_1 = 1$ , such that

$$\vec{E}_1 = \vec{p}_1 u_1(\vec{r}, \psi) \cdot e^{-i\psi}, \quad \vec{B}_1 \approx \vec{q}_1 \times \vec{E}_1, \quad \vec{p}_1 \cdot \vec{q}_1 = 0, \quad (2.iv.7)$$

where  $\vec{p}_1$  is a polarization vector ( $|\vec{p}_1| = 1$ ),  $\psi = \omega_1 t - \vec{r} \cdot \vec{k}_1$  is a retarded coordinate, and  $u_1$  is a "slow" envelope, whose variations in space  $\vec{r}$  and in  $\psi$  are much slower than  $e^{-i\psi}$  (for a *cw* plane wave,  $\vec{p}_1 u_1 = \text{const}$ ). Here  $\vec{k}_{10} = k_1 \vec{q}_{10}$ , where  $\vec{q}_{10}$  is a unity vector along the main axis of the beam propagation. Using Eqs. (2.iv.6) and (2.iv.7), we find that

$$\vec{D}^{(2)} \cdot \vec{H}^{(2)} = 0; \quad |\vec{D}^{(2)}| = |\vec{H}^{(2)}|; \quad \vec{H}^{(2)} \approx -\vec{q}_1 \times \vec{D}^{(2)}. \quad (2.iv.8)$$

The SHG wave equations can then be obtained from Eqs. (2.iv.3) and (2.iv.5) as

$$\nabla \times \vec{E}_2 + (1/c) \partial \vec{B}_2 / \partial t = 0, \quad \nabla \times \vec{B}_2 - (1/c) \partial \vec{E}_2 / \partial t = \vec{F} \cdot \chi e^{-2i\psi} \quad (2.iv.9)$$

where the *driver*

$$\vec{F} = \chi^{-1} [(1/c) \partial \vec{D}^{(2)} / \partial t + \nabla \times \vec{H}^{(2)}] \cdot e^{2i\psi} \quad (2.iv.10)$$

is a slow envelope of nonlinear driving term (or source). Using Eq. (2.iv.6) and (2.iv.7), we find

$$\vec{F} = k_1 \partial \vec{f} / \partial \psi - \nabla \times [\vec{q}_1 \times \vec{f}] + 2ik_1 \Delta \vec{q} \times [\vec{q}_1 \times \vec{f}], \quad (2.iv.11)$$

where  $\vec{\Delta q} = \vec{q}_1 - \vec{q}_{10}$  and

$$\vec{f} = \vec{p}_2 \cdot u_1^2 B_{dc}, \quad \vec{p}_2 = -4\vec{p}_1 \cdot ([\vec{q}_1 \times \vec{p}_1] \cdot \vec{e}_{dc}) + 7(\vec{p}_1 \cdot \vec{e}_{dc})[\vec{q}_1 \times \vec{p}_1]. \quad (2.iv.12)$$

with  $\vec{p}_1$ ,  $\vec{q}_1$ , and  $\vec{e}_{dc} = \vec{B}_{dc}/B_{dc}$  being the unity vectors of the wave polarization, the wave direction of propagation, and the *dc* magnetic field respectively. In Eq. (2.iv.12),  $\vec{p}_2$  is a (nonunity in general) SH polarization vector. Because of the spatial anisotropy imposed by a *dc* magnetic field, SHG depends upon the polarization of laser fundamental wave; Eq. (2.iv.12) reflects induced birefringence of the nonlinear interaction. If the fundamental wave propagates along the *dc* magnetic field  $\vec{B}_0$ , then  $\vec{D}^{(2)} = 0$ ,  $\vec{H}^{(2)} = 0$  [Eq. (2.iv.5)], and nonlinear effects vanish identically. The strongest interaction occurs when the light propagates *normally* to  $\vec{B}_0$  and is polarized *parallel* to  $\vec{B}_0$ , in which case  $\vec{p}_1 = \vec{e}_{dc}$ , and  $\vec{p}_2 = 7[\vec{q}_1 \times \vec{p}_1]$ , i. e. then the SH polarization vector,  $\vec{p}_2$ , is *normal* to the fundamental polarization vector,  $\vec{p}_1$ . This is the manifestation of birefringence. Note that when the fundamental light is polarized (and propagates) *normally* to the magnetic field, the SH polarization coincides with that of the fundamental radiation.

The first term in the rhs of (2.iv.11) is due to the time-dependence of *dc* field and the intensity of the fundamental wave, whereas the second and third terms are due to spatial nonuniformity of the wave intensity and phase respectively. Eq. (2.iv.11) clearly indicates that the driver  $\vec{F}$  (and therefore, SHG itself) vanishes (consistently with [14,15,6]), if the radiation is a *cw* plane wave and *dc* field is uniform and constant, since in such a case  $\partial/\partial\psi = 0$ ,  $\Delta q = 0$ , and  $\nabla = 0$ . Thus, the nonvanishing effect may be expected only if the field system varies in space and/or time.

We consider first the perhaps simplest and fundamental example: a *pulse* plane wave propagating in a *uniform dc* magnetic field ( $\vec{B}_{dc} = B_0 \hat{e}_z$ ;  $\vec{e}_{dc} = \hat{e}_z$ ) normal to the wave propagation axis, *y*, i. e.  $\vec{q}_1 = \hat{e}_y$ . Suppose that the amplitude (and/or phase) of fundamental plane wave is *arbitrarily* modulated in time with a (complex) envelope  $u_1(\psi)$ . SHG may occur only if the fundamental wave envelope is *time* (or  $\psi$ )-*dependent*. The plane wave solution of Eq. (2.iv.9) with a driver, Eq. (2.iv.10) is found as

$$\vec{u}_2(\psi, y) = -(\chi/2) k_1 B_0 \vec{p}_2 y d[u_1^2(\psi)]/d\psi, \quad (2.iv.13)$$

assuming that the field  $B_0$  is "turned on" at  $y=0$ . Eq. (2.iv.13) demonstrates the same dependence on the *distance* of interaction ( $u_2 \propto y$ ) as for SHG in a "classical" nonlinear medium with ideal phase-matching, with the significant difference being that SHG is proportional now to the time-derivative of the driving envelope (since  $\psi$  is a  $\psi = \omega_1 t - \vec{r} \cdot \vec{k}_{10}$  is a retarded time, see (2.iv.7)). The use of very broad spectrum radiation can further enhance the SHG effect. The efficiency of the SHG conversion of a Gaussian pulse of a full duration  $2t_p$  is evaluated as:

$$\frac{\mathcal{E}_{SHG}(y)}{\mathcal{E}_1} = \frac{1}{\sqrt{2}} \left[ \frac{\alpha p_2}{90\pi} \cdot \frac{B_0 E_1}{B_{cr}^2} \cdot \frac{y}{ct_p} \right]^2. \quad (2.iv.14)$$

Considering now *spatial* nonuniformity of nonplanar (in particular, Gaussian) wave, we assume a *cw* wave (it is clear however that a combined time/space nonuniformity may significantly enhance the nonlinear interaction), such that in Eq. (2.iv.11),  $\partial/\partial\psi = 0$ . The calculations of the beam propagation for SHG in general case become very tedious, and to make them more traceable, we consider here the simplest case of spatially nonuniform problem: the

propagation of 2-D Gaussian beam (or so called *slab* beam), in which case it is assumed uniform along only one direction (say in the  $x$ -axis, so that  $\partial/\partial x = 0$ ), and having a Gaussian profile in the other direction (say the  $z$ -axis) normal to the direction of the propagation (the  $y$ -axis). We use a paraxial approximation, which is adequate in our case. To maximize the interaction, we assume that the  $dc$  magnetic field,  $\vec{B}_{dc}$ , is normal to the direction of the wave propagation, and the fundamental wave is polarized normally to  $\vec{B}_{dc}$ , with  $\vec{E}_I = \vec{p}_I u_I e^{-i\psi}$ , where now  $\psi = \omega_I t - k_I y$  and  $\vec{p}_I = \hat{e}_x$ . A fundamental solution at the frequency  $\omega$  of Maxwell equations (2.iv.3) in such a case is a 2-D Gaussian beam:

$$u_I = E_I \cdot G(y, z); \quad G = \sqrt{Y(y)} \cdot \exp[-Y(y)z^2/2z_0^2]; \quad Y(y) \equiv (1 + iy/y_d)^{-1}, \quad (2.iv.15)$$

where  $E_I = \text{const}$  is its maximum amplitude (i. e. at the waist), a function  $G$  describes a Gaussian transverse amplitude (and phase) profile and its spatial evolution due to diffraction.  $Y(y)$  is a "diffraction" factor,  $z_0$  is the minimum size of the beam (at the waist,  $y=0$ ), and  $y_d = z_0^2 k_I = z_0/\phi_d$ , and  $\phi_d = (k_I z_0)^{-1} \ll 1$ . Here  $y_d$  is the Rayleigh parameter (diffraction length) of the beam, and  $\phi_d$  is a diffraction angle. The nonlinear driver  $\vec{F}$  (2.iv.11) is found then as:

$$\vec{F} = 4\vec{p}_I u_I^2 \left[ \frac{\partial B_{dc}}{\partial y} - i \left( \frac{B_{dc}}{y_d} \right) |Y(y)|^2 \left( 1 - |Y|^2 \frac{z^2}{z_0^2} \right) \right]. \quad (2.iv.16)$$

Here, the *first* term in square brackets is due to the *spatial nonuniformity of the dc field*, whereas the *rest of the expression* is due to the *inhomogeneity of the laser field*.

Ommitting here a tremendous amount of analytical calculations done in [3] by using driver (2.iv.16), we write here the efficiency of the SHG conversion,  $w_{SHG}(\infty)$ , for the case of a Gaussian beam in an *uniform dc* magnetic field:

$$w_{SHG}(\infty) \equiv \frac{W_{SHG}(\infty)}{W_I} = \frac{19}{16\sqrt{2}} \left( \frac{\alpha}{45} \right)^2 \left( \frac{E_I B_0}{B_{cr}^2} \right)^2 \approx 2.2 \times 10^{-8} \left( \frac{E_I B_0}{B_{cr}^2} \right)^2. \quad (2.iv.17)$$

Since  $P_{SHG}(\infty)$  is a constant independent of any parameter of the problem, one can see from Eq. (2.iv.17) that for the fixed laser power,  $W_I$ , and the  $dc$  magnetic field,  $B_0$ , the total output SHG power,  $W_{SHG}$ , is inversely proportional to the focal spot size,  $z_0$ , because  $E_I^2 \propto z_0^{-1}$ . Thus, the effect increases with tighter focusing (and vanishes for plane wave) which is perfectly expected by now.

To study the gradient-induced SHG in a *nonuniform dc* magnetic field, we considered the propagation of a Gaussian beam in the magnetic field originated by a 2-D magnetic dipole, i. e. two thin parallel "magnetic" wires [3, 107], and 4-D magnetic quadrupole [3]. As expected, both of these cases showed the generation of non-vanishing GHG. In both the cases the efficiency of the SHG conversion was directly related to the inhomogeneity of the field, and was vanishing as the dipole size was increasing for the same electrical current in the wires.

An apparent interpretation of nonvanishing SHG is that the nonuniformity allows for the momentum transfer between photons and  $dc$  field (which would ultimately result in the recoil of material system generating the  $dc$  field), thus breaking the symmetry that causes vanishing interaction of completely uniform field system. This explanation could be directly corroborated by e. g. direct QED calculations of SHG by two collinear photons + elementary source (particle) of  $dc$  field, similarly to quasi-elastic scattering of a *single* photon at a Coulomb potential [110], see also [94,108,109] (QED calculations of photon splitting probability in a nucleus Coulomb

potential using the recoil momentum can be found in Ref. [111]). Examples of such sources could be protons (or heavy nuclei) or neutrons with two collinear photons "SHG-scattered" at the particle spin and Coulomb  $dc$  (electric) field. In macroscopic terms, the paraxial approximation for SHG, Eq. (2.iv.10), is not valid here, and SHG is originated by an elementary multipole source, Eq. (2.iv.7), in a limited volume  $\ll \lambda_I^3$  (similarly to Sect. V in Ref. [98]); we found that in the lowest approximation, the source is a dipole for a spin and a quadrupole for a Coulomb field.

We did not pursue specific experimental design or optimization calculations for the SHG effect in magnetized vacuum. To do so, one needs to calculate 3-D  $cw$  (i. e. cylindric) Gaussian beam propagation in a  $dc$  magnetic field (compare with 2-D Gaussian beam here), and combine it, in general case, with temporal effect in a laser pulse (see above). However, to roughly estimate the SHG output, we made an order-of-magnitude estimate using the results [3]. In the existing lasers, the spatial compression (focusing) of laser beams is much stronger than temporal compression (i. e.  $z_0/\lambda = O(1) \ll t_p \omega/2\pi$ ). Thus, to be on conservative side, we neglected temporal effect and considered only spatial effects. Total SHG photon output,  $\Phi_{SHG}$ , was estimated then as

$$\Phi_{SHG}(s^{-1}) = \bar{P}_I w_{SHG}/2\hbar\omega \approx 1.2 \times 10^{-38} \bar{P}_I(W) I_I(W/cm^2) \lambda(\mu m) B_0^2(ts). \quad (2.iv.18)$$

where  $\bar{P}_I$  is the total time-averaged power of fundamental harmonics (in  $W$ ),  $w_{SHG}$  is the efficiency of SHG transformation (2.iv.17),  $\lambda$  is the wavelength of fundamental harmonics (in  $\mu m$ ),  $I_I$  is the maximal intensity of fundamental harmonics (in  $W/cm^2$ ) at focal point, and magnetic field  $B_0$  is in *Tesla*. Rapid advent of laser and magnet technologies makes the observation of vacuum SHG feasible in near future. Considering, for example, a laser with  $\lambda \approx 0.8 \mu m$  (as in *Ti-Spph* laser), with  $\bar{P}_I \sim 10^5 - 10^6 W$  and intensity at focal point  $I_I \sim 10^{22} W/cm^2$ , and  $B_0 \sim 10^3 ts$  (which can currently be obtained by explosions). Eq. (2.iv.18) yields then  $\sim 10^2 - 10^3$  vacuum SHG *photons/day*.

## 2.v. Preliminary new research

In his research under the reported grant, this PI has started to work on a few new prospective directions, in which he and his group and collaborators has obtained promising preliminary results. These are:

- \* Laser-powered single-particle hysteretic motional oscillator [8,9,11,12,15,22];
- \* Measuring super-short pulses with relativistic particles in a spark chamber [14,19]; and
- \* EM-bursts of the nuclear-scale time petawatt-laser-driven electrons [17,18,20,21,23].

Some of these new ideas, among other ideas, have been proposed for the new research under the current AFOSR grant.

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